

## IN THE SPECIFICATION

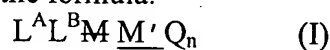
### Please amend the Specification as follows:

*At column 1, line 4, after the title and before "FIELD OF THE INVENTION" at column 1, line 5, please insert the following paragraph:*

This application is a Reissue Application of U.S. No. 09/428,576, filed October 28, 1999, previously issued as U.S. Patent No. 6,271,323.

*At column 7, line 39 carried over to column 8, line 67, please delete the paragraphs and replace them with the following paragraphs:*

In one embodiment, the bulky ligand metallocene-type catalyst compounds of the invention are represented by the formula:



where  $\underline{M} \underline{M'}$  is a metal atom from the Periodic Table of the Elements and may be a Group 3 to 12 metal or from the lanthanide or actinide series of the Periodic Table of Elements, preferably  $\underline{M} \underline{M'}$  is a Group 4, 5 or 6 transition metal, more preferably  $\underline{M} \underline{M'}$  is a Group 4 transition metal, even more preferably  $\underline{M} \underline{M'}$  is zirconium, hafnium or titanium. The bulky ligands,  $L^A$  and  $L^B$ , are open, acyclic or fused ring(s) or ring system(s) and are any ancillary ligand system, including unsubstituted or substituted, cyclopentadienyl ligands or cyclopentadienyl-type ligands, heteroatom substituted and/or heteroatom containing cyclopentadienyl-type ligands. Non-limiting examples of bulky ligands include cyclopentadienyl ligands, cyclopentaphenanthrenyl ligands, indenyl ligands, benzindenyl ligands, fluorenyl ligands, octahydrofluorenyl ligands, cyclooctatetraendiyl ligands, cyclopentacyclododecene ligands, azenyl ligands, azulene ligands, pentalene ligands, phosphoyl ligands, phosphinimine (WO 99/40125), pyrrolyl ligands, pyrozolyl ligands, carbazolyl ligands, borabenzene ligands and the like, including hydrogenated versions thereof, for example tetrahydroindenyl ligands. In one embodiment,  $L^A$  and  $L^B$  may be any other ligand structure capable of  $\eta$ -bonding to  $\underline{M} \underline{M'}$ , preferably  $\eta^3$ -bonding to  $\underline{M} \underline{M'}$  and most preferably  $\eta^5$ -bonding. In yet another embodiment, the atomic molecular weight (MW) of  $L^A$  or  $L^B$  exceeds 60 a.m.u., preferably greater than 65 a.m.u.. In another embodiment,  $L^A$  and  $L^B$  may comprise one

or more heteroatoms, for example, nitrogen, silicon, boron, germanium, sulfur and phosphorous, in combination with carbon atoms to form an open, acyclic, or preferably a fused, ring or ring system, for example, a hetero-cyclopentadienyl ancillary ligand. Other  $L^A$  and  $L^B$  bulky ligands include but are not limited to bulky amides, phosphides, alkoxides, aryloxides, imides, carbolides, borollides, porphyrins, phthalocyanines, corrins and other polyazomacrocycles. Independently, each  $L^A$  and  $L^B$  may be the same or different type of bulky ligand that is bonded to  $M$   $M'$ . In one embodiment of formula (I) only one of either  $L^A$  or  $L^B$  is present.

Independently, each  $L^A$  and  $L^B$  may be unsubstituted or substituted with a combination of substituent groups R. Non-limiting examples of substituent groups R include one or more from the group selected from hydrogen, or linear, branched alkyl radicals, or alkenyl radicals, alkynyl radicals, cycloalkyl radicals or aryl radicals, acyl radicals, aroyl radicals, alkoxy radicals, aryloxy radicals, alkylthio radicals, dialkylamino radicals, alkoxycarbonyl radicals, aryloxy carbonyl radicals, carbomoyl radicals, alkyl- or dialkyl- carbamoyl radicals, acyloxy radicals, acylamino radicals, aroylamino radicals, straight, branched or cyclic, alkylene radicals, or combination thereof. In a preferred embodiment, substituent groups R have up to 50 non-hydrogen atoms, preferably from 1 to 30 carbon, that can also be substituted with halogens or heteroatoms or the like. Non-limiting examples of alkyl substituents R include methyl, ethyl, propyl, butyl, pentyl, hexyl, cyclopentyl, cyclohexyl, benzyl or phenyl groups and the like, including all their isomers, for example tertiary butyl, isopropyl, and the like. Other hydrocarbyl radicals include fluoromethyl, fluoroethyl, difluoroethyl, iodopropyl, bromohexyl, chlorobenzyl and hydrocarbyl substituted organometalloid radicals including trimethylsilyl, trimethylgermyl, methyldiethylsilyl and the like; and halocarbyl-substituted organometalloid radicals including tris(trifluoromethyl)-silyl, methyl-bis(difluoromethyl)silyl, bromomethyldimethylgermyl and the like; and disubstituted boron radicals including dimethylboron for example; and disubstituted pnictogen radicals including dimethylamine, dimethylphosphine, diphenylamine, methylphenylphosphine, chalcogen radicals including methoxy, ethoxy, propoxy, phenoxy, methylsulfide and ethylsulfide. Non-hydrogen substituents R include the atoms carbon, silicon, boron, aluminum, nitrogen, phosphorous, oxygen, tin, sulfur, germanium and the like, including olefins such as but not limited to olefinically unsaturated substituents including vinyl-terminated ligands, for example but-3-enyl, prop-2-enyl, hex-5-enyl and the like. Also, at least two R groups, preferably two adjacent R groups, are joined to form a ring structure

having from 3 to 30 atoms selected from carbon, nitrogen, oxygen, phosphorous, silicon, germanium, aluminum, boron or a combination thereof. Also, a substituent group R group such as 1-butanyl may form a carbon sigma bond to the metal  $\text{M M}'$ .

Other ligands may be bonded to the metal  $\text{M M}'$ , such as at least one leaving group Q. For the purposes of this patent specification and appended claims the term "leaving group" is any ligand that can be abstracted from a bulky ligand metallocene-type catalyst compound to form a bulky ligand metallocene-type catalyst cation capable of polymerizing one or more olefin(s). In one embodiment, Q is a monoanionic labile ligand having a sigma-bond to  $\text{M M}'$ . Depending on the oxidation state of the metal, the value for n is 0, 1 or 2 such that formula (I) above represents a neutral bulky ligand metallocene-type catalyst compound.

*At column 9, lines 13-43, please delete the paragraphs and replace them with the following:*

In one embodiment, the bulky ligand metallocene-type catalyst compounds of the invention include those of formula (I) where  $\text{L}^{\text{A}}$  and  $\text{L}^{\text{B}}$  are bridged to each other by at least one bridging group, A, such that the formula is represented by

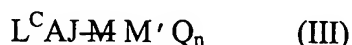


These bridged compounds represented by formula (II) are known as bridged, bulky ligand metallocene-type catalyst compounds.  $\text{L}^{\text{A}}$ ,  $\text{L}^{\text{B}}$ ,  $\text{M M}'$ , Q and n are as defined above. Non-limiting examples of bridging group A include bridging groups containing at least one Group 13 to 16 atom, often referred to as a divalent moiety such as but not limited to at least one of a carbon, oxygen, nitrogen, silicon, aluminum, boron, germanium and tin atom or a combination thereof. Preferably bridging group A contains a carbon, silicon or germanium atom, most preferably A contains at least one silicon atom or at least one carbon atom. The bridging group A may also contain substituent groups R as defined above including halogens and iron. Non-limiting examples of bridging group A may be represented by  $\text{R}'_2\text{C}$ ,  $\text{R}'_2\text{Si}$ ,  $\text{R}'_2\text{Si R}'_2\text{Si}$ ,  $\text{R}'_2\text{Ge}$ ,  $\text{R}'\text{P}$ , where R' is independently, a radical group which is hydride, hydrocarbyl, substituted hydrocarbyl, halocarbyl, substituted halocarbyl, hydrocarbyl-substituted organometalloid, halocarbyl-substituted organometalloid, disubstituted boron, disubstituted pnictogen, substituted chalcogen, or halogen or two or more R' may be joined to form a ring or ring system. In

one embodiment, the bridged, bulky ligand metallocene-type catalyst compounds of formula (II) have two or more bridging groups A (EP 664 301 B1).

*At column 10 lines 8-31, please delete the paragraphs and replace them with the following paragraphs:*

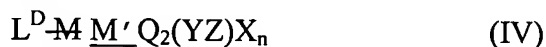
In this embodiment, the bulky ligand metallocene-type catalyst compound is represented by the formula:



where  $\underline{M \underline{M'}}$  is a Group 3 to 16 metal atom or a metal selected from the Group of actinides and lanthanides of the Periodic Table of Elements, preferably  $\underline{M \underline{M'}}$  is a Group 4 to 12 transition metal, and more preferably  $\underline{M \underline{M'}}$  is a Group 4, 5 or 6 transition metal, and most preferably  $\underline{M \underline{M'}}$  is a Group 4 transition metal in any oxidation state, especially titanium;  $L^C$  is a substituted or unsubstituted bulky ligand bonded to  $\underline{M \underline{M'}}$ ; J is bonded to  $\underline{M \underline{M'}}$ ; A is bonded to  $\underline{M \underline{M'}}$  and J; J is a heteroatom ancillary ligand; and A is a bridging group; Q is a univalent anionic ligand; and n is the integer 0, 1 or 2. In formula (III) above,  $L^C$ , A and J form a fused ring system. In an embodiment,  $L^C$  of formula (III) is as defined above for  $L^A$ , A,  $\underline{M \underline{M'}}$  and Q of formula (III) are as defined above in formula (I).

*At column 10, lines 38-67, please delete the paragraphs and replace them with the following paragraphs:*

In an embodiment, the bulky ligand metallocene-type catalyst compound is represented by the formula:



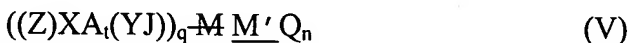
where  $\underline{M \underline{M'}}$  is a Group 3 to 16 metal, preferably a Group 4 to 12 transition metal, and most preferably a Group 4, 5 or 6 transition metal;  $L^D$  is a bulky ligand that is bonded to  $\underline{M \underline{M'}}$ ; each Q is independently bonded to  $\underline{M \underline{M'}}$  and  $Q_2(YZ)$  forms a uncharged polydentate ligand; A or Q is a univalent anionic ligand also bonded to  $\underline{M \underline{M'}}$ ; X is a univalent anionic group when n is 2 or X is a divalent anionic group when n is 1; n is 1 or 2.

In formula (IV), L and  $\underline{M \underline{M'}}$  are as defined above for formula (I). Q is as defined above for formula (I), preferably Q is selected from the group consisting of -O-, -NR-, -CR<sub>2</sub>- and -S-; Y is either C or S; Z is selected from the group

consisting of -OR, -NR<sub>2</sub>, -CR<sub>3</sub>, -SR, -SiR<sub>3</sub>, -PR<sub>2</sub>, -H, and substituted or unsubstituted aryl groups, with the proviso that when Q is -NR- then Z is selected from one of the group consisting of -OR, -NR<sub>2</sub>, -SR, -SiR<sub>3</sub>, -PR<sub>2</sub> and -H; R is selected from a group containing carbon, silicon, nitrogen, oxygen, and/or phosphorus, preferably where R is a hydrocarbon group containing from 1 to 20 carbon atoms, most preferably an alkyl, cycloalkyl, or an aryl group; n is an integer from 1 to 4, preferably 1 or 2; X is a univalent anionic group when n is 2 or X is a divalent anionic group when n is 1; preferably X is a carbamate, carboxylate, or other heteroallyl moiety described by the Q, Y and Z combination.

*At column 11, lines 25-46, please delete the paragraphs and replace them with the following paragraphs:*

In one embodiment, the bulky ligand metallocene-type catalyst compound is represented by the formula:



where  $\underline{M} \text{--} \underline{M'}$  is a metal selected from Group 3 to 13 or lanthanide and actinide series of the Periodic Table of Elements; Q is bonded to  $\underline{M} \text{--} \underline{M'}$  and each Q is a monovalent, bivalent, or trivalent anion; X and Y are bonded to  $\underline{M} \text{--} \underline{M'}$ ; one or more of X and Y are heteroatoms, preferably both X and Y are heteroatoms; Y is contained in a heterocyclic ring J, where J comprises from 2 to 50 non-hydrogen atoms, preferably 2 to 30 carbon atoms; Z is bonded to X, where Z comprises 1 to 50 non-hydrogen atoms, preferably 1 to 50 carbon atoms, preferably Z is a cyclic group containing 3 to 50 atoms, preferably 3 to 30 carbon atoms; t is 0 or 1; when t is 1, A is a bridging group joined to at least one of X, Y or J, preferably X and J; q is 1 or 2; n is an integer from 1 to 4 depending on the oxidation state of  $\underline{M} \text{--} \underline{M'}$ . In one embodiment, where X is oxygen or sulfur then Z is optional. In another embodiment, where X is nitrogen or phosphorous then Z is present. In an embodiment, Z is preferably an aryl group, more preferably a substituted aryl group.

*At column 14, after the paragraph ending at line 42 and before the paragraph starting at line 43, please insert the following paragraphs:*

The catalyst composition is prepared by forming a well-stirred suspension of finely divided filler material, one or more metallocene catalysts and one or more

cocatalysts in one or more suitable diluents, and then spray drying the suspension. Typically, in preparing the suspension, the filler material is added to a solution or dispersion of the cocatalyst to form a first suspension. The first suspension is stirred for approximately 20 to 60 minutes, and then a solution or dispersion of the metallocene catalyst is added thereto. The resulting final suspension is stirred for a further 20 to 60 minutes and then spray dried. The same or different diluents may be used for the metallocene catalyst and the cocatalyst.

Preferably, spray drying is performed by spraying the suspension through a heated nozzle into a stream of heated inert drying gas, such as nitrogen, argon, or propane to evaporate the diluent and produce solid-form particles of metallocene catalyst and cocatalyst in a matrix of filler material. The volumetric flow of the drying gas is preferably considerably larger than the volumetric flow of the suspension. Atomization of the suspension may be accomplished using an atomizing nozzle or a centrifugal high speed disc atomizer.

The diluent employed in forming the suspension is typically a material capable of dissolving or suspending the metallocene catalyst and the cocatalyst, and suspending the filler material. For example, hydrocarbons such as linear or branched alkanes including n-hexane, n-pentane and isopentane; aromatics such as toluene and xylene; and halogenated hydrocarbons such as dichloromethane are useful as the diluent. Preferably, for practical reasons the diluent has a boiling point from about 0.degree.-150.degree. C.

*At column 18, line 21, and before the paragraph at column 18, line 22, please insert the following paragraph:*

If the metal compound and/or the co-catalyst occurs naturally in liquid form, it can be introduced "neat" into the particle lean zone. More likely, the liquid catalyst is introduced into the particle lean zone as a solution (single phase, or "true solution" using a solvent to dissolve the metal compound and/or co-catalyst), an emulsion (partially dissolving the catalyst components in a solvent), suspension, dispersion, or slurry (each having at least two phases). Preferably, the liquid catalyst employed is a solution or an emulsion, most preferably a solution. As used herein, "liquid catalyst" or "liquid form"

includes neat, solution, emulsion, and dispersions of the transition metal or rare earth metal component(s) of the catalyst and/or co-catalyst.